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LABORATORY EVALUATION AND APPLICATION OF MICROWAVE ABSORPTION PROPERTIES UNDER SIMULATED CONDITIONS FOR PLANETARY ATMOSPHERES

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I. INTRODUCTION AND SUMMARY

Radio absorptivity data for planetary atmospheres obtained from spacecraft radio occultation experiments and earth-based radio astronomical observations can be used to infer abundances of microwave absorbing atmospheric constituents in those atmospheres, as long as reliable information regarding the microwave absorbing properties of potential constituents is available. The use of theoretically-derived microwave absorption properties for such atmospheric constituents, or laboratory measurements of such properties under environmental conditions which are significantly different than those of the planetary atmosphere being studied, often leads to significant misinterpretation of available opacity data. Steffes and Eshleman (1981) showed that under environmental conditions corresponding to the middle atmosphere of Venus, the microwave absorption due to atmospheric SO_2 was 50 percent greater than that calculated from Van Vleck-Weiskopff theory. Similarly, results obtained for the microwave opacity from gaseous H_2SO_4 under simulated Venus conditions, during the first two years of Grant NAGW-533, showed that not only was the opacity from H_2SO_4 much greater than theoretically predicted, but that its frequency (wavelength) dependence was far different than that theoretically predicted (Steffes, 1985 and Steffes, 1986). More recently, measurements made by Steffes and Jenkins (1987), during the third year of Grant NAGW-533, have shown that simulated Jovian conditions do indeed agree with theoretical predictions, but only at wavelengths longward of 1.3 cm. The recognition of the need to make such laboratory measurements of simulated planetary atmospheres over a range of temperatures and pressures which correspond to the altitudes probed by both radio occultation experiments and radio astronomical observations, and over a range of frequencies which correspond to those used in both radio occultation experiments and radio astronomical observations, has led to the development of a facility at Georgia Tech which is capable of making such measurements. It has been the goal of this investigation to conduct such measurements and to apply the results to a wide range of planetary observations, both spacecraft and earth-based, in order to determine the identity and abundance profiles of constituents in those planetary atmospheres.

Initially, this facility was developed, and then operated, in order to evaluate the microwave absorbing properties of gaseous sulfuric acid (H_2SO_4) under Venus atmospheric conditions. The results, obtained at 13.4 cm and 3.6 cm wavelengths, were applied to measurements from Mariner 5, Mariner 10, and Pioneer-Venus Radio Occultation experiments, to determine abundances of gaseous sulfuric acid in the Venus atmosphere, with accuracies exceeding those achieved with <u>in-situ</u> instruments (Steffes, 1985). Later efforts concentrated on making laboratory measurements of the microwave absorption from gaseous H_2SO_4 at wavelengths from 1.2 to 22 cm under simulated Venus conditions. We applied these results to radio astronomical observations of Venus which have been made in the same wavelength range, in order to better model the structure of H_2SO_4 and SO_2 abundance in the Venus atmosphere, and to resolve temporal variations of their abundances on a planet-wide basis. The results of this effort were especially rewarding in that the unique frequency and pressure dependences measured for the absorption from gaseous H_2SO_4 in these wavelength ranges has finally explained what were thought to be inconsistencies between measurements of the absorption in the Venus atmosphere at 13.3 and 3.6 cm wavelengths and those obtained in the 1 to 2 cm wavelength range. Our laboratory measurements also suggested that a substantial variation in the

Venus microwave emission, related to the abundance of gaseous sulfuric acid, might exist near the 2.2 cm wavelength. Since no observations of the Venus emission at this wavelength have ever been published, we conducted observations of Venus using the 140-foot NRAO telescope in April 1987 to not only search for the presence of the predicted feature, but to use such a feature to determine a planet-wide average for sulfuric acid vapor abundance below the main cloud layer. This observation and initial results are discussed in Section V of this report.

The highest priority activity for the first half of this grant year has continued to be laboratory measurements of the microwave properties of the simulated atmospheres of the outer planets and their satellites. As described in the previous Annual Status Report for Grant NAGW-533 (February 1, 1986 through January 31, 1987), initial laboratory measurements of the microwave opacity of gaseous ammonia (NH_3) in a hydrogen/helium (H_2/He) atmosphere, under simulated conditions for the outer planets were completed in September 1986. These measurements were conducted at frequencies from 1.3 GHz to 22 GHz (wavelengths from 1.3 cm to 22 cm), at temperatures from 178 K to 300 K, and under total pressures reaching as high as 6 atmospheres. Such measurements have long been sought by a number of researchers working on inferring ammonia abundance profiles in Jovian atmospheres. Our measurements represented the first time that measurements of the microwave absorption of gaseous ammonia under simulated conditions for the outer planets had been conducted. The results of these measurements, and their effect on the interpretation of microwave opacity data from Jupiter, obtained both from *Voyager* radio occultation measurements made at 13 cm and 3.6 cm wavelengths and from radio astronomical observations in the 1.3 cm to 20 cm wavelength range, are discussed in a paper entitled "Laboratory Measurements of the Microwave Opacity of Gaseous Ammonia (NH_3) Under Simulated Conditions for the Jovian Atmosphere," by Steffes and Jenkins, which was submitted to Icarus in December 1986, and has since been revised and accepted for publication (accepted April 20, 1987). These results showed that in the 1.38 to 18.5 cm wavelength range, the absorption from gaseous ammonia was correctly expressed by the modified Ben-Reuven lineshape as per Berge and Gulkis (1976). As a result, it became clear that the opacity which would be exhibited by a solar abundance of ammonia would be a factor of 1.5-2.0 below the opacity at the 2 to 6 Bar levels of the atmosphere as inferred from radio emission studies in the 6 to 20 cm wavelength range (see de Pater and Massie, 1985 and de Pater, 1986), and the opacity at the 1 Bar level measured by the Voyager 13 cm wavelength radio occultation experiment (see Lindal et al., 1981). This additional opacity, while most likely due to an overabundance of gaseous ammonia beyond the solar abundance, may also be partially due to other gases or condensates, especially in the 2 to 6 Bar levels of the atmosphere.

As a result, in the first half of this current grant year, we have continued such laboratory measurements of the microwave absorption and refraction from other potential microwave absorbers contained in the outer planets' atmospheres, including methane (CH_4) and water vapor (H_2O), as well as additional high-sensitivity mesurements of the absorption from gaseous NH_3 at 13.3 and 18.5 cm. It has also been brought to our attention by several researchers that significant uncertainties exist as to the actual absorption spectrum of gaseous ammonia at wavelengths shortward of 1 cm. It has been found by some (e.g., de Pater and Massie, 1985) that the observed millimeter-wave emission from Jupiter is inconsistent with the millimeter-wave absorption spectrum predicted using the modified Ben-Reuven line shape for ammonia. In order to confirm this, we have recently developed a Fabry-Perot spectrometer system capable of operation from 30 to 41 GHz (wavelengths from 7.3 to 10 mm). This system can be used at pressures up to 2 Bars and temperatures as low as 150 K, which corresponds closely to the conditions at altitudes in the Jovian atmosphere most responsible for the observed millimeter-wave absorption. Measurements are currently being conducted, and preliminary results are presented in Section IV. A complete description of the millimeter-wave spectrometer is given in Section II.

In the second half of this grant year, we hope to complete laboratory measurements of the 7.3-8.3 mm absorption spectrum of ammonia, as well as additional measurements of H_2O and CH_4. We are also investigating the feasibility of making laboratory measurements of the microwave properties of gaseous phosphine (PH_3). Like ammonia, phosphine is a symmetric-top molecule with rotational resonances at millimeter or submillimeter wavelengths. However, also like ammonia, an inversion spectrum should exist much lower in frequency (see Townes and Schawlow, 1955). Unlike the ammonia inversion spectrum, which is centered around 23 GHz, the phosphine inversion spectrum is expected below 10 MHz. However, under the proper conditions of temperature and high pressure, phosphine may exhibit measurable absorption in the 10 to 20 cm wavelength range. Laboratory measurements of the microwave properties of phosphine in a hydrogen/helium atmosphere present even greater hazards than those for the previous measurements of ammonia in a like atmosphere. Because of the high toxicity of phosphine (more than 30 times more toxic than ammonia), self-contained breathing apparatus must be available in case of accident. Likewise, special ventilation procedures will be required since phosphine can spontaneously combust. We hope to measure the microwave absorption properties of phosphine, under simulated Jovian conditions (i.e., temperatures down to 153 K and pressures to 6 atm in a hydrogen/helium atmosphere), and over the full wavelength range (1.37 cm to 18.5 cm), but only after completion of the measurements for gaseous NH_3, H_2O, and CH_4. We likewise hope to pursue a program of further analysis and application of our laboratory results to microwave data for the outer planets, such as Voyager Radio Occultation experiments and earth-based radio astronomical observations. Of equal importance, we feel however, would be the further analysis and application of our laboratory results for the microwave absorption from gaseous SO_2 and gaseous H_2SO_4 in the Venus atmosphere. Our long term goal would be a detailed analysis of available multi-spectral microwave opacity data from Venus, including data from the Pioneer-Venus Radio Occultation experiments and earth-based radio and radar astronomical observations, such as the kinds which have been performed at the NRAO Very Large Array (VLA) and at stations in the Deep Space Network (DSN). The new measurements of Venus microwave emission at 2.25 cm and 1.9 cm made with the NRAO 140-foot telescope are an especially important contribution to this data set. This would provide a chance to determine both spatial and temporal variations in the abundances of both H_2SO_4 and SO_2 in the Venus atmosphere. We intend to pursue discussions with the Pioneer-Venus Radioscience Team Leader, in order to obtain additional absorptivity data from the 3.6 and 13 cm radio occultation experiments.

II. THE GEORGIA TECH RADIO ASTRONOMY AND PROPAGATION (R.A.P.) FACILITY

The basic configuration of the planetary atmospheres simulator developed at Georgia Tech for use in measurement of the microwave absorptivity of gases under simulated conditions for planetary atmospheres is described at length in the previous Annual Status Report(s) for Grant NAGW-533. It is also discussed at length in Steffes (1986) and Steffes and Jenkins (1987). The updated simulator system, shown in Figure 1, is currently configured for simulations of the outer planets. Measurements of the microwave opacity and refractivity of test gas mixtures can be performed at frequencies from 1.6 GHz to 27 GHz (wavelengths from 1.2 cm to 18.5 cm) using two microwave resonators contained within a pressure chamber. While the pressure chamber itself is capable of containing pressures up to 10 atmospheres, and the temperature chamber is capable of achieving temperatures as low as 150 K, it has been found that the combination of high pressures and low temperatures can create a substantial problem with sealing the pressure chamber. The use of gaseous hydrogen (H_2) in the outer planets simulations has required the development of procedures and equipment for conducting such simulations. Such precautions have included the use of a hydrogen leakage sensor which is placed inside the temperature chamber immediately outside the pressure vessel (see Figure 1). This sensor can detect the potentially dangerous build-up of hydrogen gas within the freezer unit. A ventilation pump has also been provided which can be used to draw any escaping hydrogen gas out of the freezer compartment. Additional precautions have included ramps which allow all of the equipment to be moved out-of-doors to a concrete slab immediately adjacent to the laboratory. All experiments which employ gaseous hydrogen can thus be conducted out-of-doors in order to avoid any build-up of hydrogen gas within the laboratory. A covered outdoor storage area for hydrogen and helium gases has also been constructed in order to allow safe storage of these gases, and to expedite the out-of-doors experiments. In addition, two vacuum sensors are included in the system. These sensors not only allow accurate determination of pressure vessel evacuation, but they can also be used for accurately determining the abundances of microwave-absorbing test gases, which are typically very small at low temperatures, due to low saturation vapor pressures.

The overall result has been a pressure vessel large enough to contain two microwave resonators, which is capable of maintaining 6 atmospheres of pressure at a temperature of 150 K, with an acceptably small leak ratio. While the range of pressures which can be tested was not quite as large as originally hoped, the resulting range of temperatures and pressures does represent the range over which nearly all of the microwave opacity in the Jupiter atmosphere has been observed, and thus is extremely useful in interpretation of microwave opacity data from Voyager I and II radio occultation experiments, as well as from earth based radio astronomical observations, and, in the future, opacity measurements to be made using the Galileo probe. Likewise, the pressure-temperature ranges measured are close enough to those over which microwave absorption or refraction has been measured in the atmospheres of Saturn, Titan, Uranus, and Neptune, so that accurate estimates of abundances of microwave-absorbing constituents in these atmospheres can also be made.

The most recent addition to the Georgia Tech Radio Astronomy and Propagation Facility has been a Fabry-Perot type resonator capable of operation between 30 and 41 GHz. As shown in Figure 2, the resonator consists of two gold plated mirrors separated by a distance of about 20 cm. The mirrors are contained in a T-shaped glass pipe which serves as a pressure vessel capable of withstanding over 2 atm of pressure. Each of the three open ends of the pipe is sealed with an O-ring sandwiched between the lip of the glass and a flat brass plate which is bolted to an inner flange. Electromagnetic energy is coupled both to and from the resonator by twin irises located on the surface of one of the two mirrors. Two sections of WR-28 waveguide which are attached to the irises pass through the brass plate to the exterior of the pressure vessel. The end of each waveguide section is pressure-sealed by a rectangular piece of mica which is held in place by a mixture of rosin and beeswax. As shown in Figure 3, one of these ends is connected to the sweep oscillator through a waveguide section. A Ka-band mixer is attached to the other section of waveguide and is coupled to the high resolution spectrum analyzer through a calibrated section of coaxial cable. The entire resonator, including its glass pressure envelope, is placed in the temperature chamber, which is a low-temperature freezer capable of operation down to 150 K. A network of stainless steel tubing and valves connects other components such as gas storage tanks, vacuum gauges, the pressure gauge, and the vacuum pump to the resonator assembly, so that each component may be isolated from the system as necessary. When properly secured, the system is capable of containing up to two atmospheres of pressure without detectable leakage.

III. EXPERIMENTAL APPROACH

The approach used to measure the microwave absorptivity of test gases in an H_2/He atmosphere is similar to that used previously by Steffes and Jenkins (1987) for simulated Jovian atmospheres. At frequencies below 22 GHz (see

Figure 1), the absorptivity is measured by observing the effects of the introduced gas mixture on the Q, or quality factor, of two cavity resonators at particular resonances from 1.34 GHz to 21.8 GHz. At frequencies between 35 and 41 GHz, the changes in the Q of several resonances of the Fabry-Perot resonator (see Figure 3) are related to the absorptivity of the test gas mixture at these frequencies. The changes in the Q of the resonances which are induced by the introduction of an absorbing gas mixture can be monitored by the high resolution microwave spectrum analyzer, since Q is simply the ratio of the cavity resonant frequency to its half-power bandwidth. For relatively low-loss gas mixtures, the relation between the absorptivity of the gas mixture and its effect on the Q of a resonance is straightforward:

$$\alpha \approx \left(Q_L^{-1} - Q_C^{-1}\right)\pi/\lambda \tag{3}$$

where α is absorptivity of the gas mixture in Nepers km^{-1}. (Note, for example, that an attenuation constant or absorption coefficient or absorptivity of 1 Neper km^{-1} = 2 optical depths per km (or km^{-1}) = 8.686 dB km^{-1}, where the first notation is the natural form used in electrical engineering, the second is the usual form in physics and astronomy, and the third is the common (logarithmic) form. The third form is often used in order to avoid a possible factor-of-two ambiguity in meaning.) Q_L is the quality factor of the cavity resonator when the gas mixture is present, Q_C is the quality factor of the resonance in a vacuum, and λ is the wavelength (in km) of the test signal in the gas mixture.

The first experiments involved high-sensitivity measurements of the microwave absorption from gaseous NH_3 under simulated conditions for the 2 to

6 Bar pressure range of the Jovian atmosphere in the 10 to 20 cm wavelength range. These measurements were undertaken in order to help better explain the source of the microwave opacity at altitudes below the 2 atm pressure level in the Jovian atmosphere. (See Steffes and Jenkins, 1987, or the previous Annual Status Report for Grant NAGW-533, for further discussion.) These measurements were conducted at 1.62 GHz (18.5 cm) and 2.25 GHz (13.3 cm), and took advantage of special adjustments to the atmospheric simulator so as to maximize sensitivity in the 10 to 20 cm wavelength range.

Temperatures from 193 K to 300 K were used for the experiments, since lower temperatures would risk condensation of the gaseous NH_3. When the pressure vessel reached thermal stability at the desired temperature, which was monitored using both the temperature sensors and the resonant frequencies of the system, a vacuum was drawn in the pressure vessel containing the resonators, and the bandwidth and center frequency of each of resonances was then measured. A valve was then opened which allowed the ammonia gas to enter the chamber, where 17 torr NH_3 pressure was used. Measurements of the gaseous NH_3 pressure were made with the high accuracy thermocouple vacuum gauge tubes which are shown in Figure 1. Next, 5.4 atm of hydrogen (H_2) and 0.6 atm of helium (He) were added. These gases were admitted to the chamber at a sufficiently slow rate so as not to significantly affect the temperature within the chamber. The result was an atmosphere with 6 atm total pressure composed of 90 percent hydrogen, 10 percent helium, and approximately 3730 ppm ammonia. The bandwidth of each resonance was then measured and compared with its value when the chamber was evacuated in order to determine the absorptivity of the gas mixture at 6 atm total pressure. The total pressure was then reduced by venting to 4 atm, and the bandwidths were again measured.

Subsequent measurements were likewise made at 2 atm pressure. The pressure vessel was then evacuated and the bandwidths again measured so as to assure no variation (either due to thermal shift or chemical reaction) of the Q's of the evacuated resonators had occurred. The measured changes of bandwidth (Q) were then used to compute the absorptivity of the gas mixture under the various pressure conditions.

This approach has the advantage that the same gas mixture is used for the absorptivity measurements at the various pressures. Thus, even though some small uncertainty may exist as to the mixing ratio of the initial mixture, the mixing ratios at all pressures are the same, and thus the uncertainty for any derived pressure dependence is due only to the accuracy limits of the absorptivity measurements, and not to uncertainty in the mixing ratio. (This assumes that the mixing ratio is small, so that foreign-gas broadening predominates, as is the case for our measurements.) Similarly, measurements of the frequency dependence of the absorptivity from the mixture would likewise be immune to any mixing ratio uncertainty, since foreign-gas broadening predominates.

The second major set of experiments involved measurements of the microwave absorption from methane (CH_4) under simulated Jovian conditions at frequencies from 2.2 to 21.7 GHz. These experiments employed the same dual-cavity atmospheric simulator as was used for the earlier ammonia experiments, shown in Figure 1. However, since methane absorption is very low, much larger methane mixing ratios were required to achieve detectable levels of absorption at these wavelengths. Likewise, since the methane absorption is inversely proportional to temperature, the lowest possible temperatures were used so as to maximize measurable absorption. In contrast to ammonia, the saturation vapor pressure of methane is quite large, thus risk of condensation was not a consideration. As a result, absorption from pure methane at pressure up to 6 Bars and at a temperature of 153 K was measured, as well as absorption from a 40% methane, 52% hydrogen, and 8% helium atmosphere under the same conditions of temperature and pressure. These measurements were made using resonances at 2.25 GHz (13.3 cm), 8.52 GHz (3.5 cm), and 21.7 GHz (1.38 cm).

The third set of measurements were made at the same frequencies, but involved measurements of the microwave absorption of water vapor under simulated Jovian conditions. Because of the relatively low saturation vapor pressure of water, experiments were conducted at 300 K, in order to obtain sufficient water vapor for the experiment. As with the previous experiments, the dual-cavity atmospheric simulator, shown in Figure 1, was used. A major difference, however, was the source of the test gas, in this case, H_2O. Since pressurized cannisters of water vapor are not possible at room temperature or lower, a flask of liquid H_2O was used as the source of water vapor. Using techniques similar to those used by Steffes (1985 and 1986) for measurement of the microwave absorption from gaseous H_2SO_4, a precise quantity of water vapor was obtained. That is, the flask is filled with a precisely known volume of distilled water. A vacuum is then drawn in the pressure vessel containing the microwave resonantors, and the bandwidth and center frequencies of the resonances are then measured. A valve is then opened which allows the water vapor eluting from the flask to fill the evacuated pressure vessel (0.031 m^3 of open volume with the resonators in place) and reach vapor pressure equilibrium with the liquid H_2O. Note that all components which contact the water vapor are maintained at the same temperature as the flask, so as to avoid condensation.

As H_2O vapor fills the chamber, changes in the resonance center frequencies are observed. These changes are related to the H_2O vapor abundance. After approximately 10 minutes, the frequency shift ceases, as vapor pressure equilibrium is reached. The valve to the reservoir flask is then closed, and 5.4 atm of H_2 and 0.6 atm of He are admitted to the chamber containing the H_2O vapor. The bandwidth of each response is then measured and compared with its value when the chamber was evacuated in order to determine the absorptivity of the gas mixture at 6 atm total pressure. The total pressure is then reduced by venting, and the bandwidths are again measured. Subsequent measurements are likewise made at lower pressures in order to determine absorptivities at those pressures. The pressure vessel is then evacuated and the bandwidths again measured so as to assure no variation (either due to thermal shift or chemical reaction) of the Q's of the evacuated resonators has occurred. After the experiment is complete, the volume of the remaining liquid is measured and compared with the initial volume measured in order to determine the amount of H_2O vapor present in the gas mixture tested. As with the ammonia experiments, this approach has the advantage that the same gas mixture is used for the absorptivity measurements at the various pressures.

The fourth set of measurements, which are currently being conducted, involve the millimeter-wave absorption from gaseous ammonia (NH_3) in a 90% H_2/10% He atmosphere. At the present time, the Fabry-Perot resonator (shown in Figure 2) is being modified so as to allow operation at temperatures down to 150 K. Therefore, the initial measurements were conducted at room temperature. As in the previous experiments at lower frequencies, the bandwidth and center frequencies of each of four resonances (36.29 GHz, 38.43 GHz, 39.15 GHz, and 40.62 GHz) were measured in a vacuum. Next, 20 torr of gaseous ammonia is added to the system. The pressure of the ammonia gas is measured with the high-accuracy thermocouple vacuum gauge, as shown in Figure 3. Next, 1.8 atm of hydrogen (H_2) and 0.2 atm of helium (He) are added, bringing the total pressure to 2 atm. The bandwidth of each resonance is then measured and compared with its value when the chamber was evacuated in order to determine the absorptivity of the gas mixture at 2 atm total pressure. The total pressure is then reduced, by venting, to 1 atm, and the bandwidths are again measured. Finally, the pressure vessel is again evacuated and the bandwidths again measured so as to assure no variation of the Q's of the evacuated resonator has occurred. As with the previous measurements, the measured changes of bandwidths (Q's) can then be used to compute the absorptivity of the gas mixtures at each of the resonant frequencies.

In all four of the measurements described, the amount of absorption being measured is extremely small. Thus, any errors in measurements of (or other changes in) the apparent bandwidth of the resonances, not caused by the absorbing gases, could lead to significant errors in the absorption measurement. The contribution of instrumental errors and noise-induced errors on such absorptivity measurements have been discussed at length in Steffes and Jenkins (1987). However, because our latest measurements represent such small percentage changes in bandwidth, another instrumental source of error which we refer to as dielectric loading becomes a concern.

As can be seen in Figures 1 and 3, the resonators, which operate as bandpass filters, are connected to a signal source (the microwave sweep oscillator) and to a signal receiver (the high-resolution spectrum analyzer). The "Q" of the resonator, which is defined as the ratio of energy stored in the resonator to the energy lost per cycle, equals the ratio of resonant center frequency to resonance half-power bandwidth. It is not surprising, therefore, that the stronger the coupling between the resonator and the spectrum analyzer or sweep oscillator, the lower will be the Q of the resonance, since more energy will be lost per cycle through the cables or waveguide connecting the resonator to the spectrum analyzer and sweep oscillator. For this reason, we have always designed our resonators (both the coaxially-coupled cylindrical cavity resonator used below 25 GHz and the waveguide-coupled Fabry-Perot resonator used above 30 GHz) with minimal coupling, so as to maximize Q and to minimize the changes in Q that might result from changes in coupling that occur when gases are introduced into the resonators. It should be noted that these changes in coupling, which are due to the presence of the test gas mixtures, are not related to the absorptivity of the gases, but rather to the dielectric constant or permittivity of the test gas mixtures. (Hence, the term "dielectric loading.")

We have always strived to design the coupling elements of the resonators so that the changes in lossless test gas abundances (and resulting changes in dielectric constant) had little or no effect on the Q of the resonator as measured in the system. This has been no small feat in that slight imperfections in resonators, cables, coupling loops, or waveguides can make the apparent Q of the resonator appear to vary with the abundance of such lossless gases. It has now become a standard part of our experimental procedure to repeat absorption measurements for gas mixtures in which the absorbing gas is a minor constituent, <u>without the absorbing</u> gas present. For example, after measurements were made of the microwave and millimeter-wave absorption from ammonia as a minor constituent in an H_2/He atmosphere, measurements of the apparent absorption of the H_2/He atmosphere <u>without</u> the ammonia gas were made.

Since, for the pressures and wavelengths involved, the H_2/He atmosphere is essentially transparent, no absorption was expected. If any apparent absorption was detected, "dielectric loading," or a change in coupling due to the dielectric properties of the gases, was indicated.

Initially, if any evidence of dielectric loading existed, the experiments were terminated and the apparatus disassembled, including pressure seals. The cables and coupling loops were then readjusted, and the system reassembled and tested again. The entire procedure was repeated until the dielectric loading effect was eliminated or minimized. If some small variation in the resonant Q or bandwidth due to the presence of the non-absorbing gases still remained, it was added to the uncertainty or error bars for each experiment. More recently however, we have found that the effects of dielectric loading are additive, in that they add to the apparent changes of resonator bandwidth caused by the absorbing gases. Thus, as long as the effects of dielectric loading are not time variable, they can be removed by using the measured value of the Q of a resonance with the non-absorbing gases present (instead of the Q of the resonance in a vacuum) for the quantity Q_c in equation (3).

IV. RESULTS OF LABORATORY MEASUREMENTS AND THEIR APPLICATION

As described in Section III, the first experimental sequence involved measurements of the microwave absorption from gaseous ammonia (NH_3) in a 90% hydrogen/10% helium atmosphere at the frequencies 1.62 GHz (18.5 cm) and 2.25 GHz (13.3 cm). These experiments were conducted with an ammonia mixing ratio of 3730 ppm, at temperatures as low as 193 K, with total pressures up to 6 atm. The results of these measurements are shown in tabular form for 1.62 GHz (Table I) and 2.25 GHz (Table II). Note that in Table II we also include a listing of all our previous measurements of the 2.25 GHz absorption from gaseous ammonia. Figure 4 presents the results of all of our absorptivity measurements for such an ammonia mixture at 193 K. Also shown are plots of the theoretically-derived absorption spectra for such gas mixtures at these three pressures. These theoretically-derived NH_3 absorption spectra were calculated as per de Pater and Massie (1985) and Berge and Gulkis (1976), both of which employ a Ben-Reuven line shape which has been modified so as to be consistent with the laboratory results of Morris and Parsons (1970), in which the absorption from NH_3 in a high-pressure H_2/He atmosphere at room temperature and at 9.58 GHz was measured. Our theoretical spectra have been adjusted for the temperature, pressure, and mixing ratio conditions of our experiment. This mathematical expression for the absorption of ammonia was implemented in a BASIC program for which partial pressures from H_2, He, and NH_3, as well as frequency and temperature, were adjustable variables. This program was used to calculate the theoretical values for absorption given in Tables I, II, and IV and in Figures 4 and 5.

Inspection of Figure 4 shows that the laboratory data agree surprisingly well with the theoretical predictions for NH_3 opacity made using the Ben-Reuven lineshape, and are, likewise, consistent with the results of Morris and Parsons (1970). The key result of our work has been the finding that the modified Ben-Reuven lineshape appears to correctly describe the shape of the absorption spectrum from gaseous ammonia in a hydrogen/helium atmosphere in the 1.3 to 18.5 cm wavelength range even under the temperature-pressure conditions characteristic of the Jovian atmosphere. This finding answers some questions and raises others. For example, both de Pater and Massie (1985) and West et al. (1986) recognized that, based on interpretation of the Jovian emission spectrum in the 10 to 20 cm wavelength range using the theoretically-derived absorption spectrum from ammonia, opacity at pressure levels greater than 2 atm had to exceed the amount which would be caused by the solar abundance of ammonia. As a result, both sets of authors concluded that either the theoretical lineshape was incorrect and understated the opacity of ammonia at these wavelengths by a factor of 1.5-2.0, or the ammonia abundance was greater than its solar abundance by a factor of 1.5-2.0 at pressures greater than 2 atm, or that an extra opacity source, possibly H_2O condensate, was present. Thus, since our measurements of the ammonia opacity at 13.3 cm (2.25 GHz) agree quite closely with the theoretically-derived values for opacity, and since our newest results at 18.5 cm (1.62 GHz) are likewise consistent with the theoretical lineshape, it would appear that either the ammonia abundance is greater than solar by a factor of 1.5-2.0 in the deeper layers of the Jovian atmosphere, or that an extra opacity source is present.

In an attempt to resolve this question, we have made measurements of the microwave opacity of two other Jovian atmospheric constituents: methane and water vapor. The methane experiment was conducted as described in Section III, and not surprisingly, the microwave absorption was very small. Measurements were made at 153 K of the microwave absorption from a pure methane atmosphere at 6 atm pressure, and from a 40% methane, 52% hydrogen, and 8% helium atmosphere at 6 atm pressure. No absorption was measured at the 2.25 GHz (13.3 cm) or 8.52 GHz (3.5 cm) resonances. Some absorption was measured at the 21.7 GHz (1.38 cm) resonance, but its statistical significance is uncertain since the levels measured were below the error bars for the system. (For the pure methane, the measured absorption was 11.4 ± 17 dB/km. For the 40% methane mixture, the measured absorption was 9.4 ± 18 dB/km.) We intend to further study the theoretically-derived methane absorption spectrum for purposes of comparison and to determine whether further laboratory measurements at higher frequencies might be useful. However, the results strongly indicate that methane absorption in the 3-13 cm wavelength range is negligible in the 2-6 Bar pressure range of Jupiter's atmosphere, and cannot account for the additional opacity required in that altitude range.

Measurements of the microwave absorption from water vapor were conducted at 2.25 GHz, 8.52 GHz, and 21.7 GHz at 297 K in a 90% H_2/10% He atmosphere as described in Section III. A water vapor mixing ratio of 3509 ppm was used, and total pressures of 6, 4, and 2 atm were employed. None of the measurements detected statistically significant opacity. (System thresholds were 0.45 dB/km, 0.73 dB/km, and 36 dB/km at 2.25 GHz, 8.52 GHz, and 21.7 GHz, respectively.) Thus our results for the microwave opacity from water vapor (1.38 to 13.3 cm) under simulated Jovian conditions are consistent with the theoretically-based expression for opacity derived by Goodman (1969). Furthermore, our results indicate that, like methane, water vapor cannot account for the additional opacity required in the 2-6 Bar pressure range of Jupiter's atmosphere. Thus, all three of these measurements further strengthen the case for an ammonia abundance between 1.5 and 2.0 times larger than solar abundance.

The measurements of the 7.3-8.3 mm absorptivity from NH_3 in a hydrogen/helium atmosphere were conducted at 298 K as described in Section III, with an ammonia mixing ratio of 1.32×10^{-2}, at pressures of 1 and 2 Bars. An examination of the experimental results summarized in Table III and displayed in Figure 5 reveals that because of large error bars, we cannot determine whether the modified Ben-Reuven lineshape best describes the absorption profile of gaseous ammonia shortward of 1 cm. However, the consistency of our results at 2 atm suggests that the modified Ben-Reuven lineshape understates the actual absorptivity of ammonia in this range. With the exception of one measurement at 40.62 GHz, all measurements were greater than those predicted by the Ben-Reuven theory (see Table III and Figure 5). At 1 atm, however, the observed absorption is so low that no absorption was measured at several frequencies. In its present configuration, our planetary atmospheres simulator is not sensitive enough to make reliable measurements using a total pressure of 1 atm.

There are several ways that we may improve the sensitivity of the system and increase the reliability and accuracy of the measurements. The most useful is to increase the signal to noise ratio, either by increasing the quality factor of the resonator, or by increasing the amount of absorption observed. Both of these can be achieved by performing the experiment at lower temperatures. As the physical temperature of the system decreases, the surface conductivity of the gold on the resonator's mirrors increases, which improves the quality factor of the resonances. Also, as the temperature decreases, the absorption per molecule of ammonia increases rapidly since $\alpha \sim T^{-7/2}$. At 183 K, the saturation pressure of ammonia is slightly above 20 Torr. Therefore, at 183 K, the same mixing ratio at which our previous experiments were performed (1.32×10^{-2}) can be achieved. At this temperature the Ben-Reuven model predicts that the absorption will increase by a factor of 3.5. We suggest that further experiments be performed at temperatures as low as 183 K. We feel that such experiments will make it possible to better evaluate the Ben-Reuven theory in the millimeter-wave range.

V. OBSERVATIONS OF THE MICROWAVE EMISSION OF VENUS FROM 1.3 TO 3.6 cm

As discussed in Steffes (1986), our previous laboratory measurements of the microwave absorption of gaseous H_2SO_4 under simulated conditions for the Venus atmosphere suggested that the 2-3 cm Venus emission spectrum would be especially sensitive to the subcloud abundance of gaseous H_2SO_4. Since no observations of the Venus emission in this wavelength range have ever been published, we conducted observations using the NRAO 140-foot radio telescope in order to search for spectral features related to gaseous H_2SO_4 abundance. Also, since apparent temporal variations in the Venus microwave emission spectrum from 1.3 to 3.6 cm have occurred, it was felt that simultaneous measurements over that entire wavelength range would best serve our need to characterize both the magnitude and the shape of the microwave emission spectrum of Venus, with the ultimate goal of inferring abundances and distribution of the microwave absorbing constituents (predominantly SO_2 and gaseous H_2SO_4).

The observations were conducted by P. G. Steffes and J. M. Jenkins of Georgia Tech and by M. J. Klein of JPL. Observations of Venus and several calibration sources in the same approximate regions of the sky (DR21, P-2134 + 004, Jupiter, and 3C123) were made at 8.42 GHz (3.6 cm), 13.3 GHz (2.25 cm), 18.46 GHz (1.63 cm), and 22.2 GHz (1.35 cm) over a four day period from April 25 through April 28, 1987. Observations at 1.63 cm and 2.25 cm were made using the 140-foot diameter NRAO radio telescope at Greenbank, West Virginia. (The National Radio Astronomy Observatory is operated by Associated Universities, Inc. under contract with the National Science Foundation.) Simultaneous observations at 1.35 cm and 3.6 cm were made with the 64-meter diameter DSN antenna at Goldstone, California. It should also be noted that over the entire month of April 1987, daily observations of the 3.6 cm Venus emission were also made with the 34-meter diameter DSN antenna, also located at Goldstone.

The dates of the observations were selected so that Venus would be close enough to superior conjunction so that it would appear as a very small source (approximately 12 arcseconds), but still far enough from the sun to avoid interference. This was done in order to minimize difficulties with beam size correction and source size correction. The times of observation were selected so as to maximize the elevation angle of the observing antennas, and thus minimize the effects of the earth's atmosphere on the observation. Since the microwave emission spectrum of Venus is a continuum, with little narrow line structure (due to the high atmospheric pressure), wide receiver bandwidths were used, which also served to increase the accuracy of the radiometric measurement. The bandwidths used were 20 MHz (8.42 GHz), 200 MHz (13.3 GHz), 250 MHz (18.46 GHz), and 20 MHz (22.2 GHz). Receiver system noise temperatures of 81 K or less were achieved at all frequencies further increasing the sensitivity of the measurement.

Calibration and study of the observed data is ongoing and will continue during the second half of the grant year. Effects such as the variation of receiving antenna aperture efficiency with hour angle, declination, and frequency have presented challenging calibration problems, but we believe our frequent observation of reference sources will allow correction for these effects. Preliminary calibration of the data has resulted in somewhat surprising findings. "First-look" results show a relatively high Venus brightness of 655 K at 3.6 cm, which monotonically decreases with decreasing wavelength down to 520 K at 1.35 cm. This included an emission measurement of 565 K at 2.2 cm. There are two very noticeable aspects of these results. The first is the relatively high values measured for the brightness temperatures at all wavelengths. Such high values, especially at the shortest wavelengths, suggest a reduced SO_2 abundance in the atmospheric region near the clouds. At longer wavelengths (3.6 cm), the higher brightness temperatures suggest increased opacity (possibly due to SO_2) very low in the atmosphere. The second noticeable feature is the lack of the hoped-for dip in emission around 2.2 cm. A dip on the order of 50 K below the expected value was thought possible. However, since the magnitude of such a dip is directly related to the absorption coefficient of gaseous H_2SO_4 at 2.2 cm, and since the error bars on our laboratory measurement at 2.2 cm were so large (a factor of two), it is not surprising that this spectral feature was not measured. However, this will now allow us to set an upper limit on average H_2SO_4 abundance in the Venus atmosphere.

A large amount of calibration and interpretive study of these observations has still yet to be completed, but we are hoping to soon travel to JPL and meet with Dr. Klein in order to complete initial analysis and begin preparing results for publication.

VI. PUBLICATIONS AND INTERACTION WITH OTHER INVESTIGATORS

At the beginning of the current grant year, a paper was completed and accepted for publication in Icarus, describing results and applications of experiments performed during the previous year of Grant NAGW-533 (Steffes and Jenkins, 1987). This paper is described at length in Section I of this report. More recently, we have submitted summaries of our laboratory measurements for inclusion in the twentieth issue of the Newsletter of Laboratory Spectroscopy for Planetary Science. Later this year, we hope to present our most recent results for the laboratory measurements of the millimeter-wave opacity of ammonia under Jovian conditions at the 19th Annual Meeting of the Division of Planetary Sciences of the American Astronomical Society (AAS/DPS Meeting). In a related meeting being held in conjunction with the DPS meeting, entitled, "Laboratory Measurements for Planetary Science," we will present a summary of our millimeter-wave laboratory activities, and their application to the interpretation of radio absorptivity data entitled, "Laboratory Measurements of the Millimeter-Wave Absorption from Gaseous Constituents under Simulated Conditions for the Outer Planets." Also at the AAS/DPS meeting, we hope to present results of our microwave measurements of the absorptivity of H_2O and CH_4, as well as initial results from the 1.3-3.6 cm observation of Venus. Support for travel to Pasadena for these meetings will be provided by Georgia Tech in support of planetary atmospheres research. (It should also be noted that partial support for travel to NRAO for the April 1987 observation was provided by Georgia Tech.) By the end of the current grant year, we hope to be able to submit papers on these same subjects to refereed journals for publication.

In addition to the observations of Venus conducted jointly with Dr. Michael J. Klein of JPL, more informal contacts have been maintained with groups at the California Institute of Technology (Dr. Duane O. Muhleman, regarding radio astronomical measurements of Venus opacity), at the Stanford Center for Radar Astronomy (V. Eshleman, regarding Voyager results for the outer planets, and laboratory measurements), and at JPL (Drs. Michael J.

Klein, Michael Janssen, and Samuel Gulkis, regarding radio astronomical observations of the outer planets, and A. J. Kliore, regarding the Pioneer-Venus Radioscience Program). We have also been active in the review of proposals submitted to the Planetary Atmospheres Program at NASA and as a reviewer of manuscripts submitted to Icarus and the Journal of Geophysical Research, for which Dr. Steffes is an Associate Editor. Another source of close interaction with other planetary atmospheres principal investigators has been Dr. Steffes' membership in the Planetary Atmospheres Management and Operations Working Group (PAMOWG). Travel support for attendance at PAMOWG meetings has been provided by Georgia Tech.

VII. CONCLUSION

During the first half of this current grant year, we have continued to conduct laboratory measurements of the microwave properties of atmospheric gases under simulated conditions for the outer planets. A most significant addition to this effort has been our capability to make such measurements at longer millimeter-wavelengths (7–10 mm). In the second half of this grant year, we hope to complete measurements of the millimeter-wave absorption from ammonia under simulated Jovian conditions. We will also further study the feasibility of measuring the microwave and millimeter-wave properties of phosphine (PH_3) under simulated Jovian conditions, and will proceed with such measurements, if feasible. We will likewise continue to pursue further analysis and application of our laboratory results to microwave and millimeter-wave absorption data for the outer planets, such as Voyager Radio Occultation experiments and earth-based radio astronomical observations. We also intend to pursue the analysis of available multi-spectral microwave opacity data from Venus, including data from our most recent radio astronomical observations in the 1.3-3.6 cm wavelength range and Pioneer-Venus Radio Occultation measurements at 13 and 3.6 cm, using our previous laboratory measurements as an interpretive tool. The timely publication of all of these results will be a high priority.

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IX. KEY FIGURES

Table I

Absorption Summary for 1.6 GHz

Date	Temperature K	Mixing Ratio	Pressure (atm.)	α (dB/km) Measured	α (dB/km) Theoretical
9/25/86	206	3.7×10^{-3}	6.1	1.273 ± 1.0	1.35
			4.1	0.0 ± 1.0	0.66
			2.0	0.0 ± 1.0	0.18
9/17/86	193	3.7×10^{-3}	6.1	1.83 ± 1.8	1.58
			4.1	$0.92 \begin{smallmatrix}+1.3 \\ -0.9\end{smallmatrix}$	0.78
			2.0	$0.46 \begin{smallmatrix}+1.4 \\ -0.5\end{smallmatrix}$	0.21

TABLE II

Absorption Summary for 2.2 GHz

Date	Temperature K	Mixing Ratio	Pressure (atm.)	α (dB/km) Measured	α (dB/km) Theoretical
8/6/86	300	3.7×10^{-3}	6.1	0.91 ± 0.27	0.89
			4.1	$0.36^{+0.28}_{-0.27}$	0.42
			2.0	$0.27^{+0.28}_{-0.27}$	0.11
7/25/86	193	3.7×10^{-3}	6.1	1.73 ± 0.27	2.58
			4.1	0.91 ± 0.18	1.23
			2.1	0.50 ± 0.18	0.34
8/13/86	193	3.7×10^{-3}	6.1	$2.10^{+0.27}_{-0.28}$	2.58
			4.1	0.82 ± 0.23	1.23
			2.0	$0.37^{+0.18}_{-0.19}$	0.34
9/17/86	193	3.7×10^{-3}	6.1	$2.19^{+0.22}_{-0.23}$	2.58
			4.1	$1.28^{+0.22}_{-0.23}$	1.23
			2.0	$0.37^{+0.22}_{-0.23}$	0.34
7/14/86	178	4.3×10^{-4}	6.1	$0.18^{+0.28}_{-0.18}$	0.36
			3.2	$0.18^{+0.22}_{-0.13}$	0.11

Table III

Absorption Summary for Ammonia at 298K

Mixing Ratio = 1.32×10^{-2}

Date	Frequency GHz	Pressure (atm)	(dB/km) Measured	(db/km) Theoretical
5/19/87	36.29	2.0	109.3 ± 73	66.72
5/19/87	36.29	1.0	54.6 ± 73	20.13
5/20/87	36.29	2.0	127.5 ± 73	66.72
5/20/87	36.29	1.0	0.0 ± 55	20.13
5/19/87	38.43	2.0	73.0 ± 73	53.68
5/19/87	38.43	1.0	36.6 ± 73	15.43
5/20/87	38.43	2.0	54.6 ± 73	53.68
5/20/87	38.43	1.0	54.6 ± 73	15.43
5/19/87	39.15	2.0	72.9 ± 82	50.21
5/19/87	39.15	1.0	0.0 ± 82	14.22
5/20/87	39.15	2.0	72.9 ± 36	50.21
5/20/87	39.15	1.0	0.1 ± 36	14.22
5/19/87	40.62	2.0	54.8 ± 73	44.27
5/19/87	40.62	1.0	27.4 ± 73	12.24
5/20/87	40.62	2.0	18.3 ± 36	44.27
5/20/87	40.62	1.0	0.0 ± 18	12.24

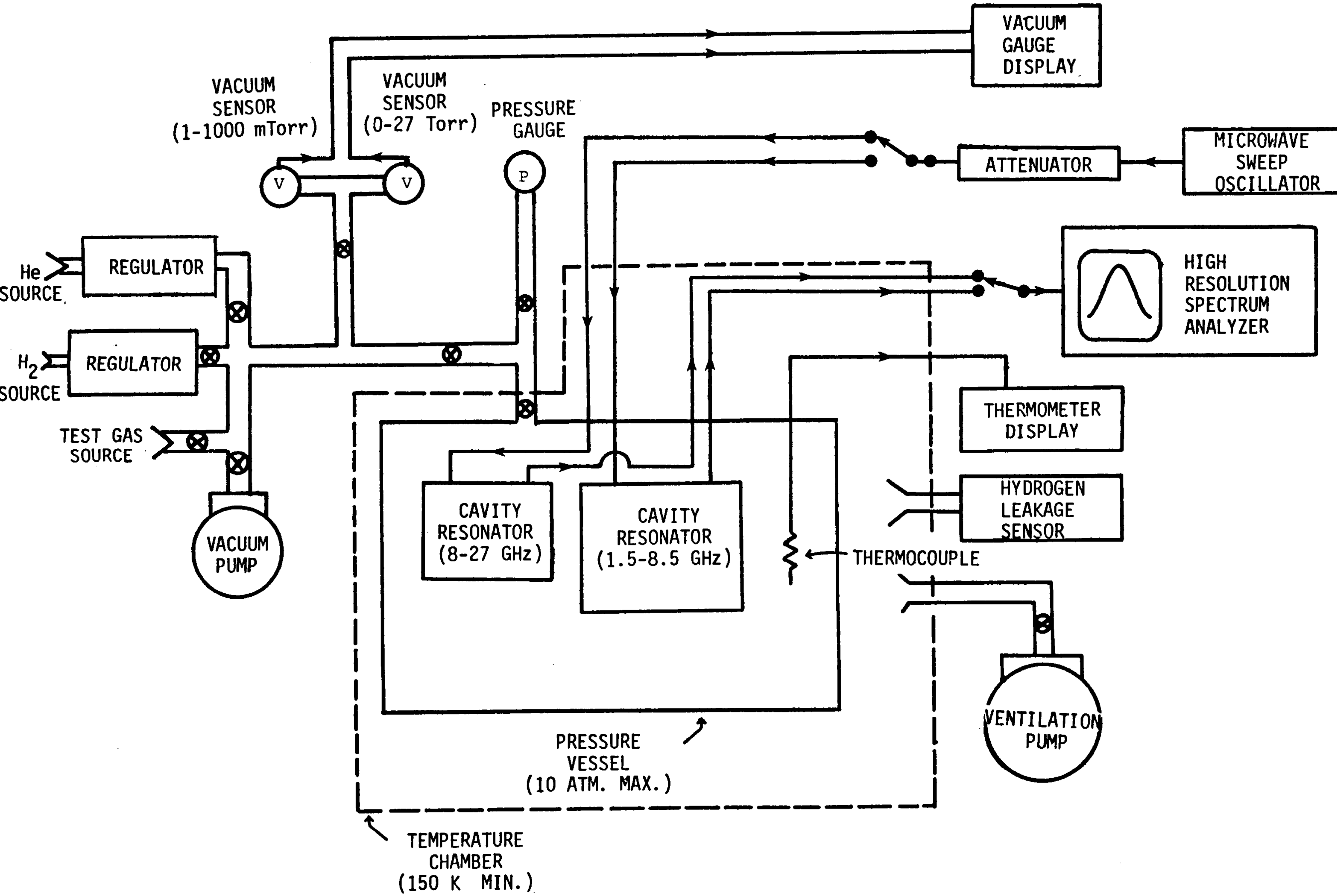

Figure 1: Block diagram of atmospheric simulator, as configured for measurements of the microwave absorption from test gases under simulated conditions for the outer planets.

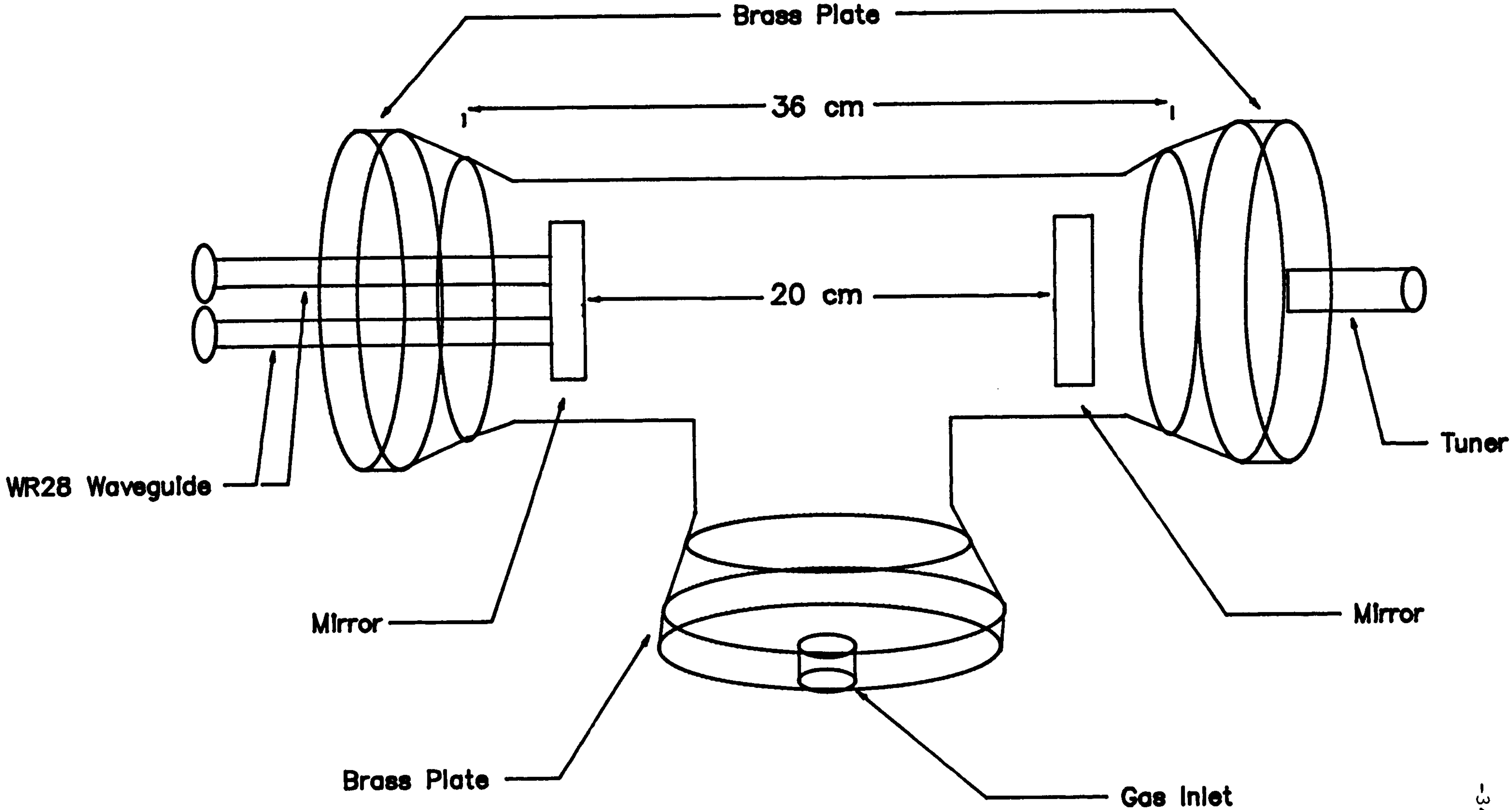

Figure 2: Diagram of Fabry—Perot Resonator

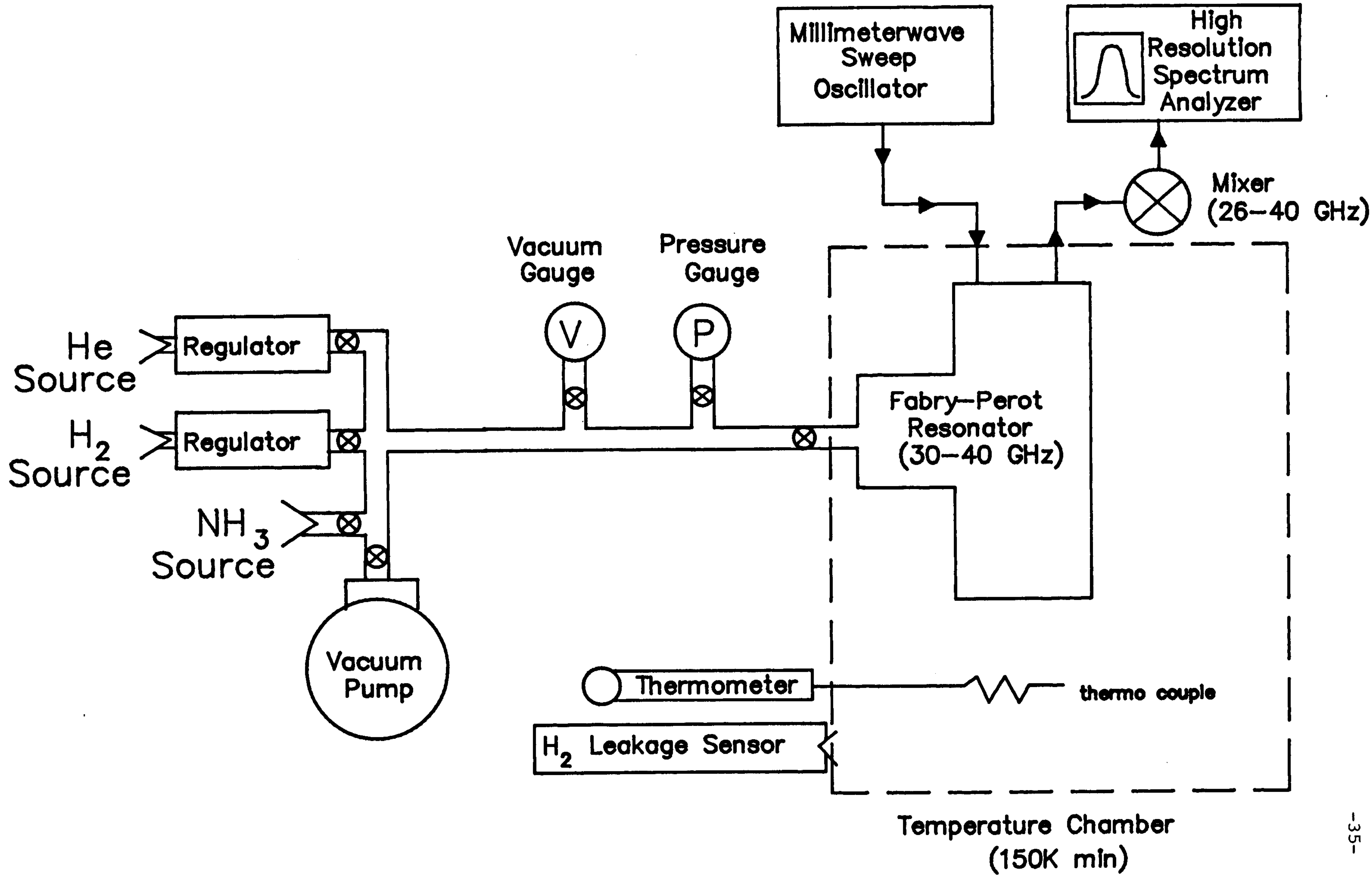

Figure 3: Block Diagram of Atmospheric Simulator

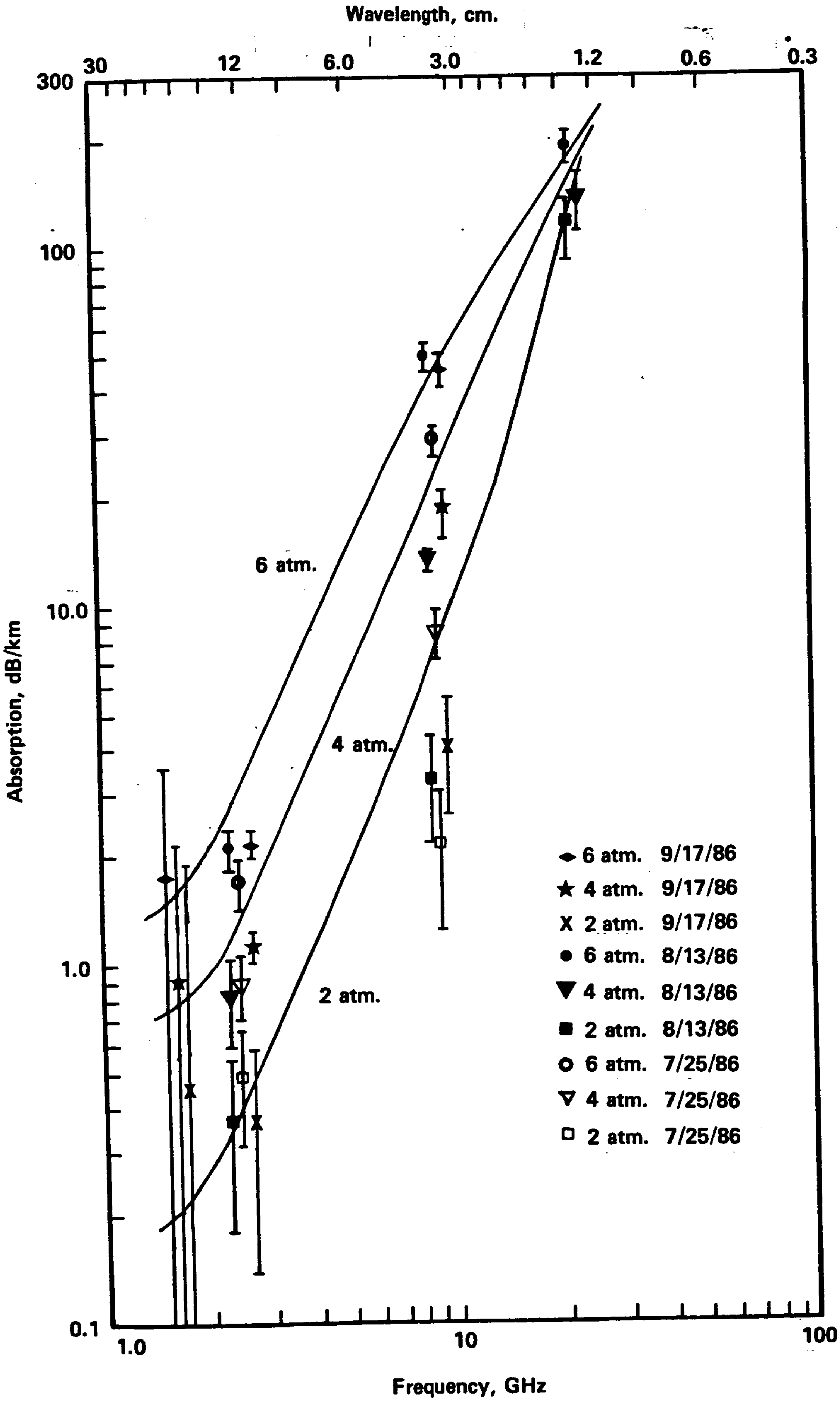

Figure 4: Measured microwave absorption from gaseous ammonia ($3.7 \pm 0.8 \times 10^{-3}$, by volume) in a 90% hydrogen/10% helium atmosphere as a function of frequency (wavelength), at 193 ± 8 K, at three different pressures. Also shown (solid lines), are theoretically derived absorption profiles.

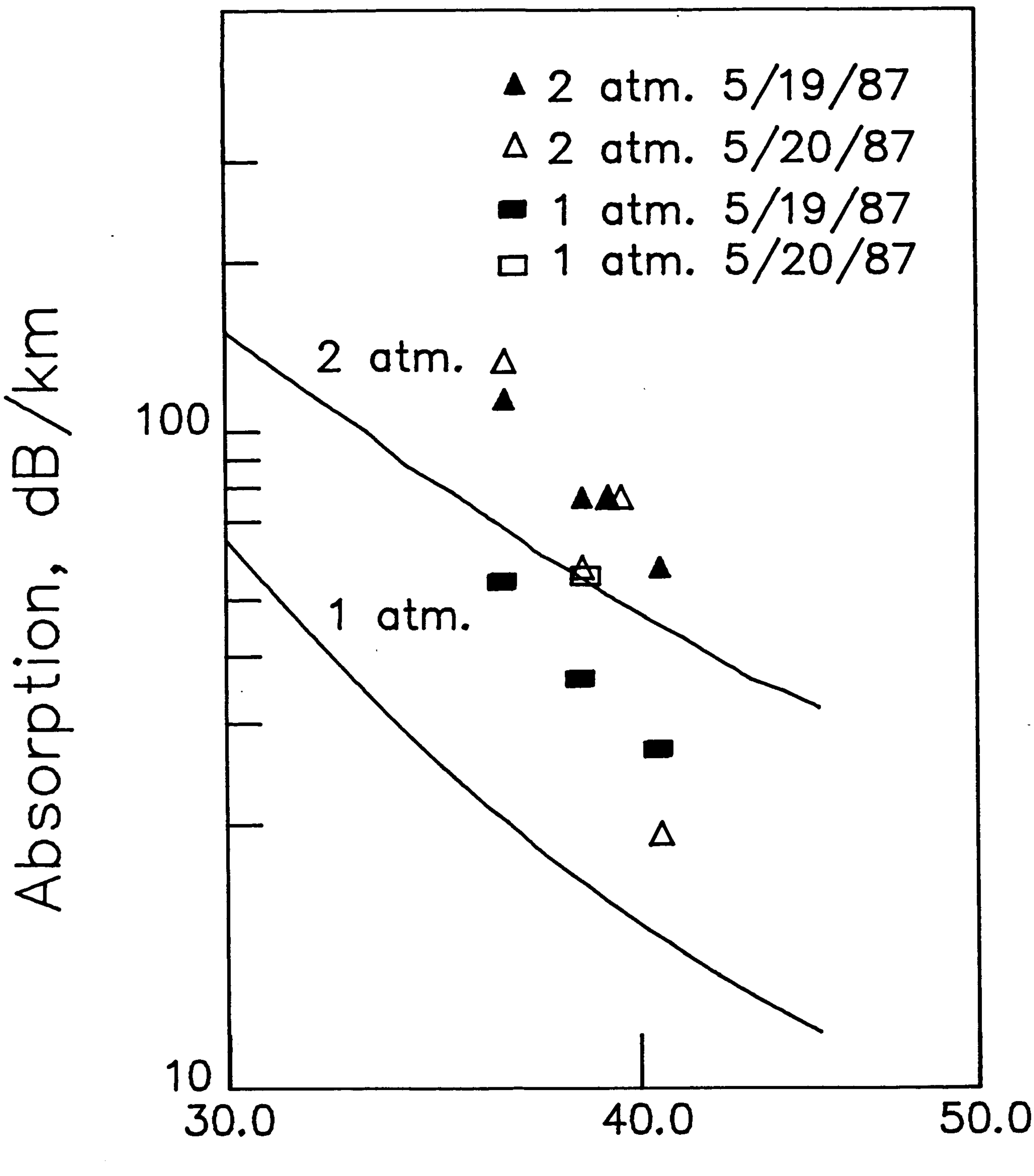

Figure 5: Graph of Theoretical and Experimental Absorption for NH_3 at Room Temperature